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Title: **OVERVIEW OF CALORIMETRIC ASSAY OF PLUTONIUM IN THE UNITED STATES**

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Los Alamos
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Overview of Calorimetric Assay of Plutonium in the United States

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Overview of Calorimetric Assay of Plutonium in the United States

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ABSTRACT

Calorimetry is a primary measurement technique for assay of quantities of plutonium in the United States. It is the most accurate NDA technique for many forms of plutonium-bearing materials. This paper provides an overview of the use of calorimetry in combination with high-resolution gamma-ray spectroscopy for accurate plutonium mass determinations.

1. INTRODUCTION

Calorimetric assay is a measurement technique that is a major component of plutonium accountability in the US. Calorimetric assay is a combination of calorimetry and isotopic analysis usually performed by high-resolution gamma-ray spectroscopy. The calorimeters used measure the total thermal power generated by plutonium inside the calorimeter measurement chamber. The thermal power is generated primarily by alpha and beta decay of plutonium isotopes, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu. Additional thermal power is generated by the isotope ^{241}Am that results from the decay of ^{241}Pu. The effective specific power, P_{eff}, is calculated from the Pu isotopic measurements that include the determination of the ^{241}Am /Pu ratio. The mass of Pu is determined by ratio of the measured thermal power determined by calorimetry, Pow, to P_{eff}, Mass (Pu)= Pow/P_{eff}. A picture of a calorimeter sensitive enough to measure high-enriched uranium as well as plutonium is shown in Figure 1. The remainder of this paper deals with the calorimetry part of calorimetric assay.

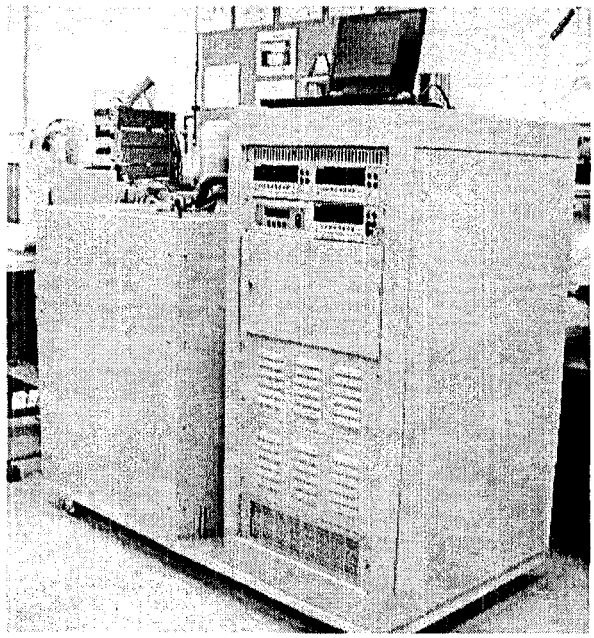

Figure 1. Radiometric calorimeter

2. HISTORY

The first calorimetry measurements related to the quantitative measurements of plutonium first took place in 1943 at the Monsanto Laboratory located in Dayton, Ohio. At that laboratory calorimetry measurements of ^{210}Po were performed. Calorimetry research and development was continued at Mound laboratory from 1948 – 1996. 250 calorimeters were constructed during that period of time for use at Mound and other US locations for the assay of nuclear material. During this period of time Mound was the lead laboratory for the development of radiometric calorimetry. Other US national laboratories and facilities were also involved in calorimetry development and research. Mound was also involved in the construction of ^{238}Pu heat sources used for electrical power generation. The ^{238}Pu heat sources could also be calibrated against traceable electrical standards and then used as secondary standards for the calibration of calorimeters. Approximately 275 encapsulated ^{238}Pu heat standards with a power range of 0.0001 to 115 Watts were built and calibrated using standards calorimeters with electrical components traceable to the US National Institute of standards and Technology. They started to be used widely in 1965. No new heat standards have been fabricated since 1992. Periodically, every 3 years, the ^{238}Pu heat standards were sent back to the Mound heat standards laboratory and recalibrated, often with different standards calorimeters and analysts. A comparison of replicate measurements made over a time period of up to 30 years provides an estimate of the uncertainty of the power value. A summary of heat standard performance from replicate measurements made during 1965 – 1996 is shown in Figure 1. For thermal powers between 0.1 W and 10 W the replicate measurements for individual standards vary less than 0.01% RSD. In the 1970s the development of high-resolution Ge detectors provided the isotopic determination capability that made calorimetric assay of plutonium a complete NDA technique. Throughout this time and up

to now the USDOE has provided the funding and impetus to develop the calorimetric assay technique in the US.

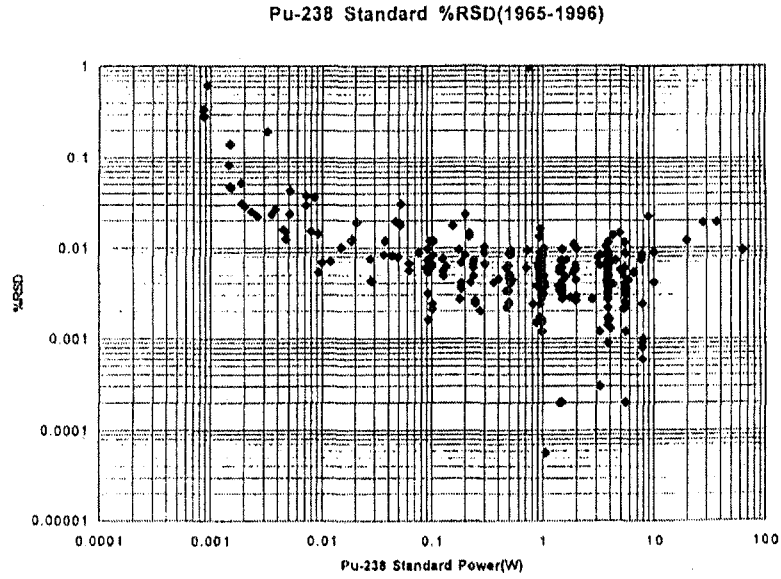

Figure 2. ^{238}Pu heat standard uncertainties determined by repeat measurements made during 1965 –1996.

3. CALORIMETER PERFORMANCE

The reason that calorimetric assay is used extensively for Pu accountability is that the technique is accurate, essentially bias free, relatively immune to sample matrix and geometry effects, and requires no representative physical standards. An example of the accuracy and bias observed with calorimeters is shown in Figure 2. In this Figure is shown the bias and precision for two 10" diameter twin-bridge calorimeter observed in an R&D facility over a one-year period by repeated measurements of different ^{238}Pu heat standards. The precision changes from 0.5% RSD at 1 W to 0.1% RSD at 12 W. The overall bias is not significantly different from 0.0%. The matrix independence is due to the fact that the magnitude of the heat flux leaving the container at equilibrium is not affected by the matrix. An example of this is shown in Figure 3 where a calorimeter response to a 1-W ^{238}Pu heat standard measured with different matrix materials is plotted. This graph shows that although the time to reach thermal equilibrium depends on the matrix, the magnitude of the signal is the same at equilibrium. Measurement times can range from 20 min to 24 hours depending on the matrix, mode of calorimeter operation, and whether temperature of the item to be measured was set close to the final equilibrium temperature using preconditioning thermostats. Some facilities have installed multiple calorimeters, up to 27 in one facility, to overcome the relatively long measurement times. Calorimeters are designed so that the total heat flux generated by the item being assayed flows through arrays of heat sensors such as resistance thermometers or thermopiles. Since 100% of the heat flux is measured, the position of the heat source inside the

calorimeter doesn't affect the result. An example of this is shown in Figure 4 that demonstrates the insensitivity of the equilibrium response of two different calorimeters to different vertical positions of a heat source.

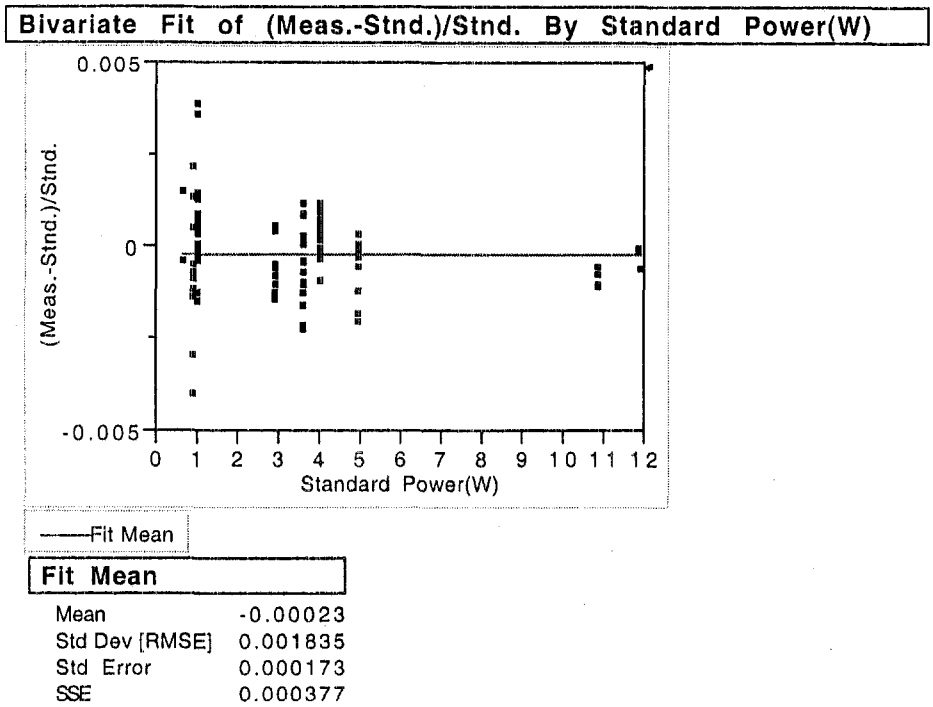

Figure 3. Precision and bias estimates measured in 1998 for two 10" twin-bridge calorimeters.

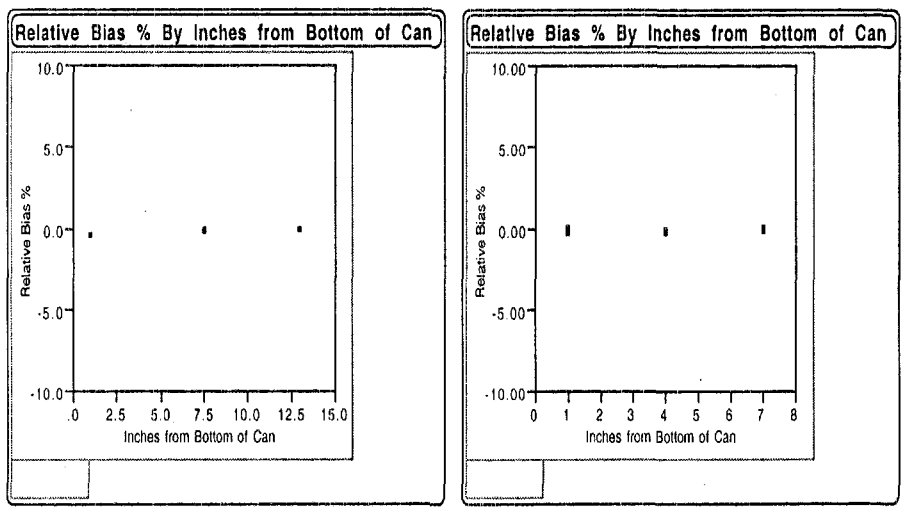

Figure 4. Vertical position sensitivity for two calorimeters

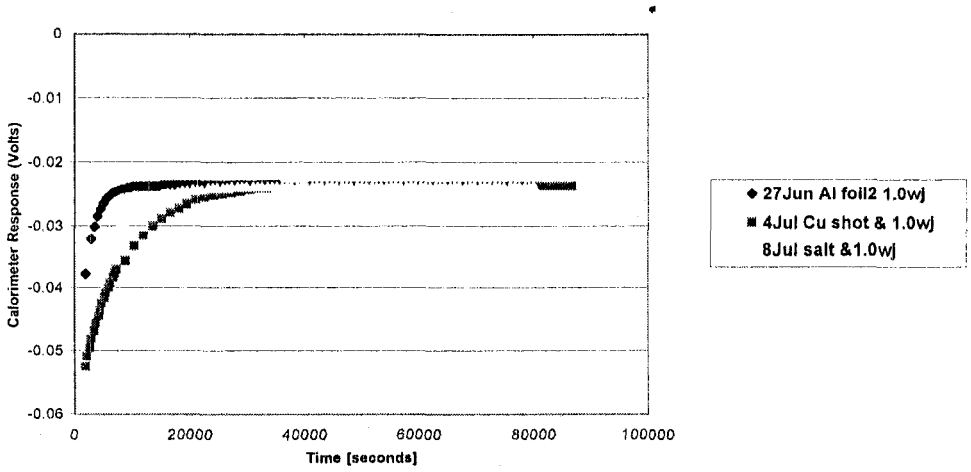

Figure 5. Calorimeter response to 1-W ^{238}Pu heat standard in different matrices.

3. CALORIMETRY EXCHANGE PROGRAM

A calorimetry exchange (CALEX) program involving the measurement of well-characterized 400 gram PuO_2 items, has been operational for 20 years in the US. Identical CALEX items were prepared and distributed to a number of US facilities for measurement. In this program each facility determines the thermal power by calorimetry and Pu isotopic composition by gamma-ray spectroscopy and reports the replicate results to the USDOE New Brunswick Laboratory (NBL). The results of the measurement are published annually. A summary of the calorimetry power bias and precision results are shown for each facility in Tables 1 and 2[1]. In Table 1 we see that the reported annual biases are typically less than 0.2%, while in Table 2 the precisions are mostly less than 0.3% RSD.

Table 1. Power measurement bias summary, USDOE/NBL Calorimetry Exchange, 400 gram Pu oxide items, 6% ^{240}Pu. [1]

Year	LLNL	LANL	Mound	Hanford	RF-CSL	RF-AL	SRP
1990	0.09	-0.11	-0.02	-0.05	0.59	0.32	
1991	0.19	0.06	-0.09	-0.10		0.12	
1992	-0.16	0.06	-0.05	-0.08	0.21	0.13	
1993	0.65	0.16	0.05	-0.04		0.24	
1994		0.19	0.05	-0.13		0.24	
1995	-0.04	0.18		-0.05			
1996	0.12	0.11		-0.11		0.19	
1997	-0.06	0.04				0.10	
1998		0.03		-0.09		0.01	-1.19
1999	0.12	-0.09			-0.05	0.00	-0.68

Table 2. Power measurement precision summary, 400 gram Pu oxide items, 6% ^{240}Pu. USDOE/NBL Calorimetry Exchange [1]

Year	LLNL	LANL	Mound	Hanford	RF-CSL	RF-AL	SRP
1990	0.13	0.53	0.12	0.28	0.20		
1991	1.36	0.22	0.18	0.23		0.16	
1992	1.30	0.15	0.18	0.24	0.37	0.12	
1993	1.01	0.22	0.16	0.28		0.20	
1994		0.18	0.20	0.30		0.20	
1995	0.22	0.13		0.29			
1996	0.15	0.16		0.39		0.40	
1997	0.08	0.22				0.20	
1998		0.21		0.35		0.17	0.72
1999		0.55		0.30	1.14	0.32	

4.WORKING STANDARDS and VERIFICATION

The accuracy and matrix independence of calorimetric assay has led to its use as a method to estimate the biases of other, more rapid NDA techniques and to produce working physical standards for verification campaigns. Two examples of the use of calorimetry as providing a reference value to check the performance of other NDA techniques are given in Figures 6 and 7. In Figure 6 is plotted a comparison of the results of calorimetry/isotopic assay vs tomographic gamma-ray scanning of plutonium contained in 7" diameter cans containing pyrochemical salts consisting of a mixture of Pu, Am, Mg, KCl, NaCl, and $MgCl_2$ [2]. The TGS random error and bias was estimated assuming the 94 calorimeter results were very close to the true value. The total measurement error for TGS assay of pyrochemical waste in cans was estimated to be 9%. A comparison of cal/iso results versus passive neutron coincidence counting (PNCC) results for a range of Pu masses in scrap and waste with non-hydrogenous matrices is shown in Figure 7. The variability of the neutron results is due to matrix variations rather than counting statistics [3].

Transportable calorimeters and high resolution Ge detectors were used by Mound Laboratory personnel to support USDOE safeguard audits by verifying portions of plutonium inventories at different US facilities from 1980 to 1995. The equipment was shipped to the facility, set-up, and used to measure items selected by the auditors. The length of the measurement campaign was two weeks involving two persons from Mound. Item measurement time with a 9" diameter, servo-controlled calorimeter ranged from 4 to 8 hours and the total number of items measured ranged from 15 to 31 items selected from 1 or 2 material categories. In the 1980s a passive neutron coincidence counter (NCC) was added to the suite of instruments to provide a higher throughput. The calorimetry/isotopic measurement was used to establish standards for the NCC. The results of Mound calorimeter verification measurements made at Los Alamos National Laboratory (LANL)in 1994 on items containing plutonium oxide are shown in Figure 8. The calorimeter results agree with the book values within 1% for most items [4].

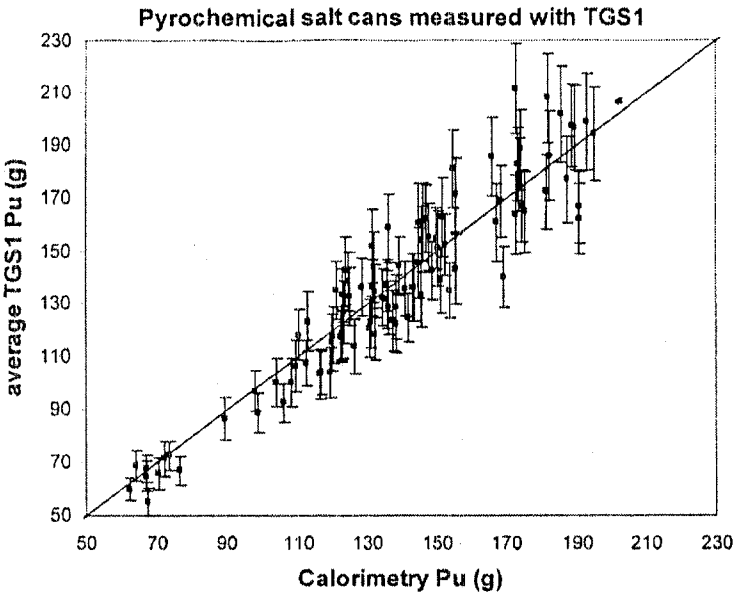

Figure 6. Average TGS measurements for the total plutonium contained in pyrochemical salt waste cans versus the corresponding calorimetric assay values [2].

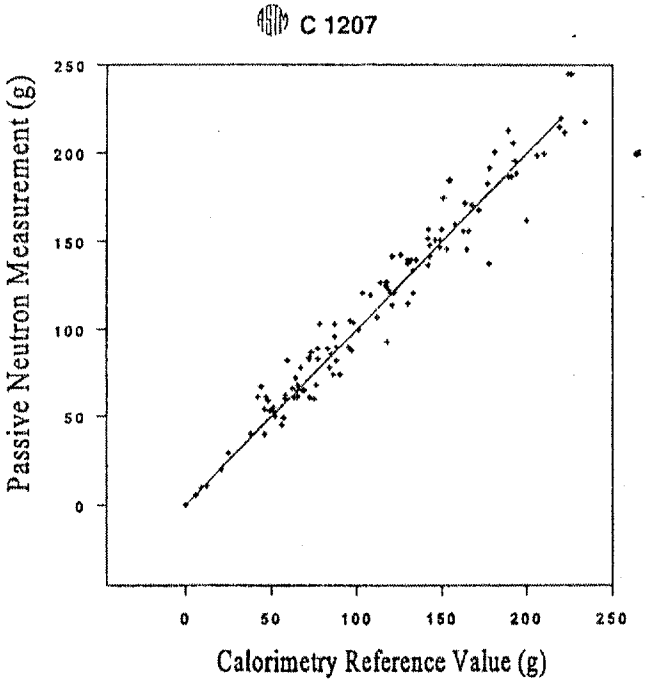

Figure 7. Comparison of PNCC results with calorimetric assay results [3]. (Figure reproduced with permission of ASTM)

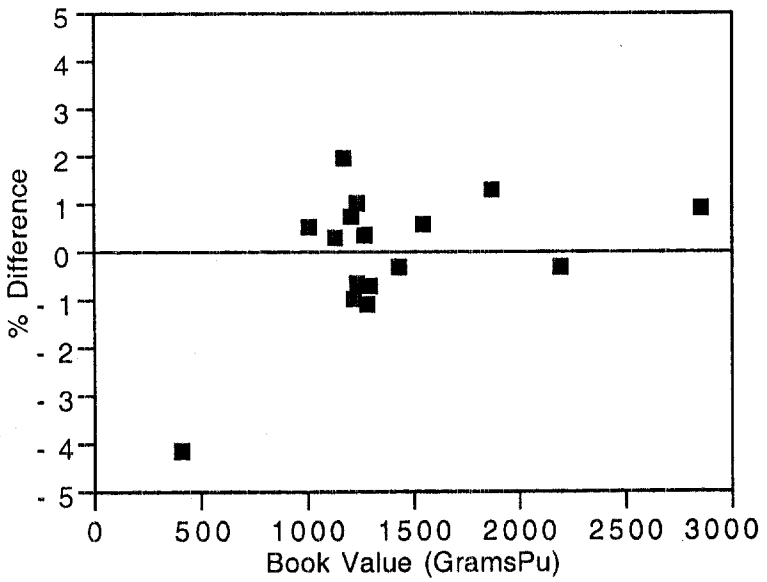

Figure 8. Mound calorimetric verification results on low-fired plutonium oxide inventory items at LANL [4].

5. CONCLUSION

This paper has given a short summary of the capabilities of calorimetric assay that has made it the dominant NDA technique for the measurement of large quantities of plutonium. A more detailed overview of calorimetric assay as practiced in the US may be found in reference 5.

6. REFERENCES

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